

## **REMARKS/ARGUMENTS**

Claims 1-12, 14-36 and 39-43 are pending in this application. These claims have all been rejected.

In response to the present Office Action, claim 1 is amended to further distinguish the claimed effect pigments from the cited prior art by further specifying the at least one metal chalcogenide as an “intrinsically colored” metal chalcogenide. Support for this amendment is found at p. 9, lines 10-13 and 31-35 of the application as filed.

Additionally, claims 14 and 16 are canceled without prejudice or disclaimer. The cancellation of claim 16, however, required a corresponding amendment to change the dependency of claim 17 from claim 16 to claim 1.

Still further, claim 18 is amended to clarify the language of that claim, as well as to change the dependency of the claim from claim 14 (now canceled) to claim 1.

Finally, claim 31 is amended to delete from the subject claim the recitation of the preferred range of from 15 to 55% by weight. New claim 44 has, therefore, been added to the application to recite the preferred range originally set forth in claim 31.

All of the claim amendments and the new claim discussed above are entirely supported by the application as originally filed. Thus they raise no issue of new matter and their entry into the file of the application is respectfully requested. Upon such entry, claims 1-12, 15, 17-36 and 39-44 will be pending in the application.

### **Objections to the Claims**

Claim 18 is objected to because of informalities in the language contained in the claim, as set forth on p. 2 of the Office Action.

Applicants have, therefore, amended claim 18 in a manner that is believed to overcome the objection. The Examiner is thus respectfully requested to reconsider and withdraw the objection to claim 18.

### **Claim Rejections Under 35 U.S.C. §112**

Claims 16-17 are rejected under 35 U.S.C. 112, second paragraph, due to the fact that claim 16 depends upon claim 13, which was previously canceled.

In response to the rejection, claim 16 is canceled without prejudice or disclaimer. Thus, claim 17 which previously depended from claim 16, has been amended to change its dependency to claim 1. The above amendments are believed to overcome the rejection of claims 16-17.

Furthermore, claim 31 is also rejected under 35 U.S.C. 112, second paragraph, due to the fact that the claim recites both a broad range and a narrower, preferred range. In response to this ground of rejection, applicants have amended the subject claim to delete the narrow range, i.e., of from 15 to 55% by weight. Additionally, new claim 44, which recites the narrow range deleted from claim 31, is proposed for addition to the application. The above described amendments are believed to overcome the ground for rejection of claim 31.

The Examiner is, therefore, respectfully requested to reconsider and withdraw the rejections of applicants' claims under 35 U.S.C. 112, second paragraph.

#### **Claim Rejections Under 35 U.S.C. 103**

At p. 4 of the Office Action, claims 1-12, 14-17, 19-36 and 39-43 are rejected under 35 U.S.C. 103 over USP 5,964,936 to Reisser, in view of USP5,624,486 to Schmid et al., as evidenced by USP 6,648,957 to Andes et al., based on the reasons set forth at pp. 4-15 of the Office Action. The rejection is respectfully traversed by the applicants for the reasons provided herein.

In contrast to the subject matter claimed in the present application, Reisser (US 5,964,936 A) advises one skilled in the art to use oxidized aluminum or aluminum alloy pigments as a base pigment for additional coloring with inorganic or organic color pigments by coating the base pigment with a color pigment-bearing metal oxide layer (see Reisser, column 3, lines 8 to 12; column 6, lines 46 to 50; claim 12).

Furthermore, Reisser additionally teaches that:

*"By superimposing the colour of the oxidized aluminium pigments with the colour of the pigments additionally applied to the pigment surfaces, that gives pigments with new colour effects and a level of colour intensity which cannot be obtained by*

*colouring conventional aluminium pigments with colour pigments."*

(see Reisser, column 6, lines 50 to 55).

Thus, it is apparent to one skilled in this art that the teaching of Reisser is directed to the generation of an absorption color, which may be altered by superimposing the absorption color of the base pigment with the absorption color of a color pigment-bearing metal oxide layer. However, an absorption color does not generate a color flop, since the resulting color impression of an absorption color does not depend upon the angle of incidence of the light.

Moreover, it is also noted that the oxidized aluminum or aluminum alloy pigments described by Reisser do not generate a color flop since a single layer of aluminum oxide/hydroxide on an aluminum or aluminum alloy core can never be responsible for a color flop.

In this respect, the Examiner is respectfully referred to Comparative Example 7 of the present application, which is the oxidized aluminum pigment of Example 1 without subsequent coating with iron oxide (see, e.g., the present application, page 18, lines 15 and 16), that is sold under the Trademark Aloxal® 3010 by the Applicant. The data providing a more detailed characterization of the pigment of Comparative Example 7 is listed in Table 2 on page 20 of the present application.

The colorimetric properties and covering power parameters at  $L^*_{110^\circ, \text{white}}/L^*_{110^\circ, \text{black}} = 0.9$  are depicted in Table 3 on page 24 of the present application, whereas the colorimetric data for the characterization of the color flop of the pigment of Comparative Example 7 compared to the pigments according to the present invention is depicted in Table 4 on page 27 of the present application.

As indicated by the subjective impression of the coloristics, as depicted in Table 3, the pigment of Comparative Example 7 does not show a color flop, which is further substantiated by the data presented in Table 4.

In order to observe a color flop of a pigment application, it must possess a certain degree of color saturation, i.e., chroma  $C^*$ . In the case of uncolored applications, although a color location  $H^*$  can be measured and determined, the observer does not observe this color at all.

Accordingly, a color flop and in particular a weak color flop also cannot be observed (see the present application at page 10, lines 17 to 20).

Thus, even though a color location  $H^*$  can be measured for the pigment of Comparative Example 7, the markedly reduced color saturation, as indicated by the chroma  $C^*$ , prevents the observation of this color at all. Accordingly, it is clearly apparent from the data depicted in Tables 4, that the oxidized aluminum or aluminum alloy pigments shows almost no color flop.

On the other hand, embedding a color pigment into a subsequently applied metal oxide layer, as recited in Reisser, i.e. in column 3, lines 8 to 12, causes only an absorption color due to absorption and scattering of light at the discrete color pigment particles in the subsequently applied metal oxide layer.

The color pigment-bearing metal oxide layer is produced by the controlled hydrolysis of metal acid esters in the presence of color pigments in an organic solvent by means of a sol-gel process which is carried out in one step. The metal acid ester is hydrolyzed and then condensed, while finally metal oxides are formed in a sol-gel process, which metal oxides are deposited on the aluminum flakes in the form of a dense closed coating and, in that case, simultaneously fix the color pigments on the metal surface. The color pigments are discrete particles in a metal oxide layer, whereas in the present application the oxidized aluminum pigments or aluminum alloy pigments exhibit at least one highly refractive metal chalcogenide layer being an intrinsically colored metal chalcogenide layer. The intrinsically colored metal chalcogenide layer effects the interference color of the resulting pigment, whereas discrete particles in a metal oxide layer only cause an absorption color. With increasing layer thickness the color caused by absorption of light by color pigments would appear more intense, whereas the color caused by interference of light, changes.

Contrary to the teaching of Reisser, applicants' pigment as presently claimed exhibits at least one highly refractive metal chalcogenide layer having a refractive index of  $>1.95$ , said at least one highly refractive metal chalcogenide layer being an intrinsically colored metal chalcogenide layer. The intrinsically colored metal chalcogenide layer of the pigment of the present invention is capable of generating an interference effect resulting in an angle dependent interference color and a color flop (as also recited in, e.g., claim 1), as indicated by the data

depicted in Tables 3 and 4 of the present application.

Thus, in contrast to the assertions regarding Reisser as set forth in the Office Action, the subject reference neither teaches the application of an intrinsically colored metal chalcogenide layer having a refractive index of  $>1.95$  onto the oxidized aluminum or aluminum alloy pigments nor the provision of effect pigments capable of generating a color flop.

Turning next to a discussion of the Schmid '486 reference, in contrast to what is claimed by applicants in the present application, one of ordinary skill in the relevant art is advised by Schmid et al. (US 5,624,486) to apply a second layer (B) consisting essentially of metal and/or nonselectively absorbing metal oxide onto the first layer (A) consisting *inter alia* of aluminum oxide and/or aluminum oxide hydrate (see Schmid et al., Abstract). These layers are applied on the substrate one after another, resulting in luster pigments having discrete layers based on a plate like substrate.

According to the teaching of Schmid et al., the first layer (A) is applied by a wet chemical coating process of the substrate with aluminum oxide/hydroxide by hydrolytic decomposition of organic aluminum compounds in the presence of an organic solvent.

As stated in ¶8 to ¶10 of Dr. Henglein's declaration dated April 29, 2009 the wet-chemical coating process disclosed in Schmid et al. and the chemical wet-process oxidation technique used in forming the pigments claimed in the present application are two unrelated methods.

The organic aluminum compounds disclosed in Schmid et al., column 6, lines 22 to 28 are known for their fast and complete hydrolysis reaction in the presence of water resulting in the consumption of water and the formation of aluminum oxide/hydroxide. As a by-product of the hydrolysis reaction, an alcohol is released.

Under the preferred conditions specified by Schmid et al. in column 6, lines 3 to 48, the hydrolysis reaction of organic aluminum compounds allows the formation of a relatively thick layer in form of an unbroken, interference-capable film (see Schmid et al., column 7, lines 10 to 21), which optical properties are defined by the deposited material.

In contrast to the wet-chemical coating process disclosed in Schmid et al., the chemical wet-process oxidation used (and claimed) in the present application is characterized by a prolonged exposure of the substrate or core to water without the simultaneous deposition of metal

oxides or metal hydroxides on the aluminum substrate or core resulting from the hydrolysis of externally supplied organic metal compounds. This process leads to a partial conversion of the substrate to aluminum oxide/hydroxide, e.g., the aluminum oxide on the aluminum substrate is produced by a direct oxidation of the aluminum substrate used.

As stated in the present application on page 11, lines 36 to 39, the pigments as claimed by applicants consequently do not exhibit a coating produced by deposition of aluminum oxide/hydroxide as is conventionally applied to pigments, but the production of an aluminum oxide/hydroxide layer from within and on the aluminum pigment or aluminum alloy pigment.

Under the conditions specified by the present application, the chemical wet-process oxidation of the metallic core, e.g., the aluminum core, results in the growth of the aluminum oxide/hydroxide layer in part into the core and in part on the core (see page 8, lines 10 to 12 of the present application). Due to the simultaneous liberation of hydrogen gas, the aluminum oxide/hydroxide layer becomes porous, allowing water to penetrate the aluminum oxide/hydroxide layer and to react with the unoxidized aluminum substrate. As this proceeds, the metallic core becomes severely roughened and structured comprising dendritic elevations and depressions.

Submitted with this Amendment is a Third 37 C.F.R. Declaration of Dr. Frank Henglein which responds to several of the Examiner's objections (as set forth on p. 17 of the present Office Action) to the declarant's previously submitted declarations, and which additionally provides further evidence tending to demonstrate the non-obviousness of pigments formed with the use of a chemical wet process oxidation technique (according to the claims of this application), in comparison to pigments made using the 'prior art' wet chemical coating process, i.e., as taught in the Schmid et al. '486 patent. Entry and consideration of the new Henglein declaration is respectfully solicited.

When comparing the Scanning Electron Micrograph (SEM) image provided with the declaration of Dr. Henglein dated September 16, 2008 (see Exhibit A) and the image of the pigment produced in Comparative Example 6 (see Dr. Henglein's Third Declaration, e.g., at ¶4), which was obtained according to the wet chemical coating process described in Schmid et al. by depositing an aluminum oxide/hydroxide layer on the aluminum substrate resulting from the

hydrolysis of externally supplied organic aluminum compounds, e.g. aluminum triisopropylate, the structural differences between the two pigments are readily apparent.

The surface of the chemical wet-process oxidized aluminum pigment (cf. declaration of Dr. Henglein dated September 16, 2008, Exhibit A) is roughened and structured comprising dendritic elevations and depressions. This particular structure is very unique and always obtained by the chemical wet-process oxidation.

The wet-chemically  $\text{Al}_2\text{O}_3$ -coated aluminum pigment (Comparative Example 6), in contrast, has a rather smooth surface without any roughening as the chemical wet-process oxidized aluminum pigment.

Advantages associated with this process related feature are e.g. the reduction in thickness of the aluminum core by the ingrowing aluminum oxide or hydroxide, which likewise reduces the total layer thickness of the resulting pigment. Thus, the relative covering power of the pigment is increased and a better orientation and stacking of the pigment in the application medium is obtained (see page 12, lines 8 to 14 and line 32 to 37 of the present application). The chemical wet-process oxidation causes, moreover, an extremely strong composite system between the aluminum oxide/hydroxide layer and the aluminum core, accompanied by a strong mechanical stability of the final effect pigment (cf. page 12, lines 1 to 6 of the present application).

The subsequently applied metal chalcogenide coating on the aluminum oxide/hydroxide layer produced as recited in the present claims exhibits a very good adhesion and forms a mixed layer with the aluminum oxide/hydroxide layer. This mixed layer is another process related characteristic that does not exist in pigments as disclosed in Schmid et al.. Due to the roughened surface of the aluminum oxide/hydroxide layer the subsequently deposited metal chalcogenide coating penetrates at least partially into the pores present in the aluminum oxide/hydroxide layer and is securely anchored therein. This anchorage is always stronger than that of external layers deposited on an aluminum core having a smooth surface (see page 12, lines 8 to 14 of the present application).

The production-related fatty acids adhering to the surface of an aluminum pigment represent a natural hydrophobic sealing layer, which prevents firm anchorage of an external layer deposited on the aluminum or aluminum alloy core (see page 12, lines 8 to 11 of the present

application). Furthermore the anchorage of the subsequently deposited coatings on a smooth surface could not be compared to that on a roughened surface.

Further to the above, the structure of the aluminum oxide/hydroxide layer has a direct effect on the color flop properties of the finally coated pigment (see page 8, lines 13 to 16 of the present application).

In contrast to the pigments as claimed by applicants, which desirably possess a soft color flop or an intense color with no substantial color flop (cf. page 5, lines 13 to 18 of the present application), it is an object of Schmid et al. to provide particularly strong metallic effect pigments (see Schmid et al., column 2, lines 37 to 39), which possess a particularly pronounced color play (see also, Schmid et al., column 4, lines 12 to 14) and a hue that for instance flops into the complementary color in the second or third interference series (Schmid et al., column 4, lines 19 to 25).

The pigments disclosed by Schmid et al. are characterized by a relatively thick layer (A) in form of an unbroken, interference-capable film (see Schmid et al., column 7, lines 10 to 21) and which essentially determines the hue of the pigments (see Schmid et al., column 4, lines 10 to 15). Thus, a uniform and optical homogenous layer results in a maximized interference effect and a particularly pronounced color play.

In contrast, the porous structure of the aluminum oxide layer obtained by the chemical wet-process oxidation used to form applicants' claimed pigments, causes the subsequently applied metal chalcogenide layer to be deposited at least partially in the pores. Thus, a definite mixed oxide layer is formed resulting in a gradient of the refractive index within the layer package along the perpendicular to the pigment surface, which results in the soft color flop of the effect pigment of present invention (see page 8, line 36 to page 9, line 4 of the present application).

Due to the chemical wet-process oxidation used/claimed by applicants, the thickness of the aluminum core is significantly reduced - resulting in a reduced total layer thickness of the pigment of the present application. The reduced total layer thickness improves the orientation and the stacking of the pigment in the application medium resulting in an increased luster, decreased gray haze and better distinctness of image (see page 12, lines 16 to 37 of the present application).

As stated in ¶13 of Dr. Henglein's declaration dated April 29, 2009 the teaching of Schmid



et al. advises the skilled person to apply a second layer (B) consisting essentially of metal and/or nonselectively absorbing metal oxide onto the first layer (A).

In contrast to the teaching of Schmid et al. the pigment of the present invention exhibits at least one highly refractive metal chalcogenide layer having a refractive index of  $> 1.95$ , said least one highly refractive metal chalcogenide layer being an intrinsically colored metal chalcogenide layer (as now recited in, e.g., amended claim 1).

An intrinsically colored metal chalcogenide layer selectively absorbs the light impinging on the effect pigment. Thus, the intrinsically colored metal chalcogenide layer is a selectively absorbing metal chalcogenide layer.

Summarizing the above arguments, one having at least an ordinary level of skill in the relevant art would not find any incentive in the prior art cited by the Examiner to modify the process for producing pigments disclosed therein in order to obtain effect pigments with a soft color flop, as presently recited in applicants' claims.

In fact, in contrast to the conclusion reached by the Examiner, applicants submit that in order to obtain a layer (A) which is in form of an unbroken, interference-capable film a person of ordinary skill would take precautions to prevent the liberation of hydrogen during the deposition of layer (A). For example, Schmid et al. advises the skilled person that in case the organic silicon and/or aluminum compounds is metered in continuously at elevated temperature the water is preferably not included in the initial charge but likewise metered in continuously (see Schmid et al., column 6, lines 49 to 63) in order to prevent an uncontrolled oxidation of the aluminum substrate.

Thus, the person skilled in the art would not be let toward combining the teaching of Reisser and Schmid et al. in order to obtain interference-capable pigments.

Furthermore, the cited state of art does not suggest to the skilled person that the porous structure of the aluminum oxide layer and the definite mixed oxide layer formed between the aluminum oxide layer and the subsequently applied metal chalcogenide layer is linked with the advanced optical properties described in the present application.

Thus, one of ordinary skill in the art does not find any suggestion in the cited references that applying at least one intrinsically colored highly refractive metal chalcogenide layer having a

refractive index of  $> 1.95$  onto an aluminum oxide-containing or aluminum oxide/hydroxide-containing layer obtained by chemical wet-process oxidation results in an effect pigment with a weak color flop having a  $DH^*_{\text{anchor}}$  values of from 1.5 to 50 as recited in applicants' claim 1.

Nothing in the cited references teaches or suggests to the skilled person the pigments of the present invention, which possess a soft color flop or an intense color with no substantial color flop. Therefore, the presently claimed pigments clearly involve an inventive step.

Further to the above, even if an individual having ordinary skill in this art did contemplate to combine the teaching of Reisser and Schmid et al., such individual certainly would not arrive at the pigments as recited in applicants' claims. Instead, such person would apply a second layer (B), that according to the teaching of Schmid et al. consists essentially of metal and/or nonselectively absorbing metal oxide, onto the first layer (A) obtained by the process described by the teaching of Reisser. Certainly, the skilled person does not get any incentive from the cited references to apply at least one highly refractive metal chalcogenide layer having a refractive index of  $>1.95$ , said at least one highly refractive metal chalcogenide layer being an intrinsically colored metal chalcogenide layer onto an aluminum oxide-containing or aluminum oxide/hydroxide-containing layer obtained by chemical wet-process oxidation.

The Andes et al. '957 reference, which constitutes the third member of the triumvirate of references relied upon by the Examiner to reject applicants' claims, certainly does not supply the elements of the claimed invention missing from the Reisser and Schmid references discussed above. Andes et al. is cited, it appears, simply due to its disclosure that magnetite is a high refractive index material which is known to have a refractive index higher than 1.8 (Office Action, p. 8). The reference is additionally cited in the paragraph bridging pp. 10-11 of the Office Action as disclosing that  $\text{SiO}_2$  and  $\text{Al}_2\text{O}_3$  are both considered low refractive index materials having a refractive index in the range of 1.35 - 1.8. None of the disclosure contained in Andes et al., however, when taken in conjunction with the combined disclosure of Reisser and Schmid, would teach or even suggest applicants' claimed pigments to one having an ordinary level of skill in this art, i.e., based on the reasons presented above.

Based on the claim amendments and the arguments and evidence (see, e.g., the Third Declaration of Dr. Henglein) presented herein, the Examiner is respectfully requested to

reconsider and withdraw the rejection of claims 1-12, 14-17, 19-36 and 39-43 under 35 USC 103(a).

Further to the above, claim 18 is separately rejected under 35 USC 103(a) on p. 15 of the Office Action as being allegedly unpatentable over Reisser in view of Schmid et al., as evidenced by Andes et al. and as further evidenced by U.S. Patent No. 5,763,086 to Schmid et al. This rejection is also respectfully traversed.

Claim 18, as amended, depends upon claim 1 and, as such, the subject claim includes all of the features recited in the independent 'parent' claim. For the reasons presented above, claim 18 is, thus, believed to be distinguishable over the combination of Reisser, Schmidt et al. '486 and Andes et al.

According to the Office Action (see p. 16), the above-cited combination does not disclose the use of zirconium oxide, tin oxide, zinc oxide or titanium oxide as the layer coating the metal oxide layer having a low refractive index. The Examiner thus has combined Schmidt et al '486 with the other three references as containing the 'missing' disclosure.

Applicants respectfully submit, however, that even the inclusion of the additional disclosure offered by the combination of Schmidt et al. '486 to that of the other three cited references would not teach or even suggest the pigments as presently claimed by applicants for the reasons presented above in the discussion of the rejection under 35 USC 103(a) of claims 1-12, 14-17, 19-36 and 39-43, which are expressly incorporated by reference into this discussion concerning the rejection of claim 18.

For the above reasons, therefore, the Examiner is respectfully requested also to reconsider and withdraw the rejection of claim 18.

### **Response to Arguments**

At pp. 17-18 of the Office Action, the Examiner sets forth her Response to the arguments set forth by applicants in the previous Amendment dated April 29, 2009 and to the evidence contained in the first and second Declaration(s) Under 37 C.F.R. §1.132 of co-inventor Dr. Frank Henglein. Objections 1-3 are responded to in Dr. Henglein's supplemental, i.e., Third Declaration which is submitted with this Amendment.

The Examiner notes in her objection #4, however, that no indication as to the roughness of applicants' pigments is set forth in the claims of the application. In response, applicants submit that a close review of the application as filed indicates that there are no numerical values contained in the written description which would permit them to assign a mathematical value to the amount or degree of roughness which their pigments display. Since it was thus not possible to quantify the pigment roughness, applicants have written the claim (i.e., claim 1) using a product-by-process format wherein it is stated that the surface of the claimed pigments is wet-chemically oxidized. It is well known by those having at least an ordinary level of skill in this art, i.e., as indicated in Dr. Henglein's previously filed declarations under 37 C.F.R. 1.132 that the wet chemical oxidation process practiced and claimed by applicants will inherently, i.e., necessarily, result in the formation of a roughened surface on the pigment, whereas the methodology taught by the Schmid et al. reference would, also necessarily, result in the production of a smooth surface. The appearance of both of these 'types' of surfaces, and the differences in the properties accorded to pigments having the respective surfaces, is explained in detail in the evidentiary declarations of Dr. Henglein that have been, and are being, submitted in this application.

M.P.E.P. §2113, headed "Product-by-Process Claims" states, in pertinent part, "The structure implied by the process steps should be considered when assessing the patentability of product-by-process claims over the prior art, especially where the product can only be defined by the process steps by which the product is made, or where the manufacturing process steps would be expected to impart distinctive structural characteristics to the final product." [citing *In re Garnero*, 412 F.2d 276, 279; 162 USPQ 221, 223 (CCPA 1979)]. The above-described situation is exactly the situation faced by applicants in the case of the present application, i.e., the surface 'roughness' can only be defined by the methodology by which the product is made - there is no numerical or other value available for comparison between the invention and the prior art - and the manufacturing steps recited in, e.g., applicants' claim 1 would be expected to impart distinctive structural properties, i.e., a roughened surface, to the surface of the subject pigments.

Applicants, therefore, respectfully submit that they have, in fact, made the only possible indication of the roughness of the claimed pigments available to them under the circumstances by

making use of the "product-by-process" option in their claim(s) as submitted to the Examiner. The Examiner should, therefore, withdraw the objection to Dr. Henglein's prior declaration(s).

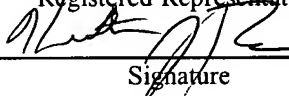
### Summary

The amendments made to applicants' claims, taken together with the additional evidence presented in Dr. Henglein's Third Declaration Under 37 C.F.R. §1.132 and the arguments provided by applicants' attorneys, are believed by applicants to overcome all of the grounds for rejection raised by the Examiner. The Examiner, therefore, is respectfully requested to reconsider and withdraw all of her objections and rejections of the claims and to issue a Notice of Allowance for this application.

I hereby certify that this correspondence is being deposited with the United States Postal Service as First Class Mail in an envelope addressed to: Commissioner for Patents, P.O. Box 1450, Alexandria, VA 22313-1450, on October 7, 2009.

Keith J. Barkaus

Name of applicant, assignee or  
Registered Representative

  
Signature

October 7, 2009

Date of Signature

Respectfully submitted,



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making use of the "product-by-process" option in their claim(s) as submitted to the Examiner. The Examiner should, therefore, withdraw the objection to Dr. Henglein's prior declaration(s).

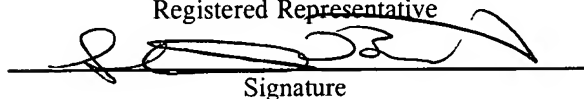
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\_\_\_\_\_  
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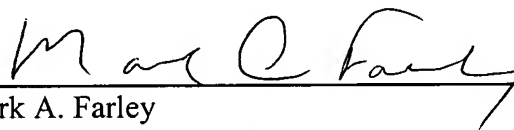
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